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Distribution and sequential extraction of some heavy metals in urban soils of Guiyang City, China

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Abstract Sixty-two soil samples collected from different functional zones of Guiyang were analyzed for total concentrations and sequential extraction of Cr, Cu, Pb, Zn and Cd by ICP spectrometry. The average total concentrations of Cr, Cu, Pb, Zn and Cd in the soils of Guiyang were 92.9, 51.6, 44.1, 139.3 and 0.28 mg/kg, respectively. The soils have been polluted by Cr, Cu, Pb, Zn and Cd to some extent in comparison with the background values of Guiyang. Significant differences were recognized in the concentrations of Cr, Cu, Pb, Zn and Cd in different functional zones. As for the sequential extraction, Cr, Cu and Zn were present mainly in the residual fraction, and Pb was present mainly in the oxidizable fraction. The reducible fraction of Cd accounts for 47.5%, and the residual fraction is lowest. The mobility and bioavailability of heavy metals follow the order of Cd>Pb>Cu>Cr>Zn.

Key words soil; heavy metal; speciation; Guiyang City

1 Introduction

The mobilization of heavy metals into the biosphere due to human activities has become an important process in the geochemical cycling of those metals (Chen Tongbin et al., 2005). This is acutely evident in urban areas where various stationary and mobile sources release large quantities of heavy metals into the atmosphere and soil, exceeding the natural emission rates (Nriagu, 1989; Bilos et al., 2001).

Urban soils are known to have peculiar characteristics such as unpredictable layering, poor structure, and high concentrations of trace elements (Kabata-Pendias and Pendias, 1992; Tiller, 1992). They are the ‘recipients’ of large amounts of heavy metals from a variety of sources including industrial wastes, vehicle emissions, coal burning waste, and others. Furthermore, any contamination of urban soils could cause in turn the accumulation of heavy metals in the food chain (Li Hongyan et al., 2006) and groundwater contamination because metals of the polluted soils tend to be more mobile than those of unpolluted ones (Steinmann and Stille, 1997; Wilcke et al., 1998). Total analysis may give information about the possible enrichment of heavy metals in soils, but it is generally recognized that it is the chemical forms of metals in the soils that determine their mobilization capacities and behaviors in the environment (Lu Ying et al., 2003). The mobility of trace metals, as well as their bioavailability and related eco-toxicity to plants, depends strongly on their specific chemical forms or ways of binding (Quevauviller et al., 1997). Sequential extraction provides information about the differentiation of the relative bonding strength of metals on various solid phases and about their potential reactivity under different physicochemical environmental conditions. And sequential extraction is considered useful for evaluating the mobility and bioavailability of heavy metals in urban soils (Sonia Pénilla et al., 2005; Şenol Kartal et al., 2006; Jeffrey et al., 2005). Many different sequential extraction procedures have been developed and used to define operationally the speciation of trace metals in matrices such as sediments and soils (Rauret et al., 1988; Maiz et al., 2000; Katherine and Christine, 2003). And one of them is the BCR method (Ure et al., 1993), which has been widely adopted by a large group of specialists.

The objective of this study is: (1) to evaluate the contamination of Cr, Cu, Cr, Pb and Zn in surface soil in Guiyang City; (2) to determine the chemical speciation of heavy metals in the soil using a sequential procedure in order to assess mobility and availability to plants.

2 Materials and methods

2.1 Description of the study area

Guiyang, the capital of Guizhou Province, is located nearly 1050 m above sea level (Hu Jian et al., 2006). Guiyang City lies between east longitude 106°07′ to 106°17′ and north latitude 26°11′ to 27°22′ (Wang Ji et al., 2006), and the total proper covers an area of 8084 km^2 and the population 1.56 million. It features an undulating topography, with high mountains and plateaus from north to south, and hills and river valleys in the centre. It has a subtropical monsoon climate with an annual average temperature of 15.6℃, relative humidity of 77%, and an average annual rainfall of 1177 mm. Soils in the Guiyang area are developed on widely distributed bedrocks of dolomite and limestone. Northeasterly wind is prevailing in winter, and southerly wind in summer (Guiyang Monitoring Station, 2001).

2.2 Soil collection

Surface soil samples were taken at 0–10-cm depths by means of a shovel from 62 locations in the urbanized areas (Fig. 1). About 1.0 kg of sample was taken from each sampling location, put into polyethylene bags and transported to the laboratory. The soil samples were air dried and ground to pass a 0.154 mm (100 meshes) stainless-steel sieve.

2.3 Analytical methods

Particle size analysis was made using the pipette method (Gee and Bauder, 1986). Organic C was measured using Walkley-Black titration (Nelson and Sommers, 1982).

The concentrations of Cr, Cu, Pb, Zn and Cd in the soils were determined by the triple acid digestion method as described by Lu Rukun (1999). About 0.1 g of soil was digested in a triple acid mixture of $HClO_4$, HNO_3 and HF. The solution was transferred into acid-cleaned Teflon beakers and dried on a heating plate. To completely remove fluoride ions, 1 mL $HClO_4$ was added twice to the dried residue and evaporated to incipient dryness. The final residue was diluted to a volume of 50 mL with 1 mL HNO_3.

The speciation of heavy metals present in the soil samples was evaluated using the BCR sequential extraction procedure (Ure et al., 1993), and the procedure is described below.

2.3.1 Step I: easily soluble

40 mL of 0.11 M acetic acid was added in 1 g of soil (as received) in an 100-mL centrifuge tube and extracted by shaking for 16 hours at ambient temperature (overnight). No delay should occur between the addition of the extractant solution and the beginning of the shaking. The extract was separated from the solid residue by centrifugation and decantation of the supernatant liquid into a high pressure polyethylene container. The container was stoppered and the extract was analyzed immediately or stored at 4℃ prior to analysis. The residue was washed by adding 20 mL of distilled water, shaking for 15 minutes and centrifuging. The supernatant and discard were decanted, and care was taken not to discard any of the solid residues. The 'cake' was broken, which was obtained upon centrifugation by using a vibrating rod prior to the next step.

Fig. 1. Map showing the sampling sites in Guiyang.

2.3.2 Step II: reducible

40 mL of 0.1 M hydroxylamine hydrochloride was added in the residue from Step I in the centrifuge tube and extracted by shaking for 16 hours at ambient temperature (overnight). No delay should occur between the addition of the extractant solution and the beginning of the shaking. The extract was separated from the solid residue by centrifugation and decantation as in Step I. The extract was retained in a stoppered polyethylene tube, as before, for analysis. The residue was washed by adding 20 mL of distilled water, shaking for 15 minutes, and centrifuging. The supernatant liquid and discard were decanted, and care was taken to avoid discarding any of the solid residues. The residue was retained for Step III and the 'cake' was broken, which was obtained upon centrifugation

with a vibrating rod prior to the next step.

2.3.3 Step III: oxidizable

10 mL of 8.8 M hydrogen peroxide solution was carefully added, in small aliquots to avoid losses due to violent reaction, in the residue in the centrifuge tube. The vessel was covered with a watch glass and digested at room temperature for 1 hour with occasional manual shaking. The digestion continued for 1 hour at 85℃ and the volume was reduced to a few mL by further heating of the uncovered vessel in a steam bath or equivalent. Another 10 mL of 8.8 M hydrogen peroxide was added and the covered vessel was heated again to 85℃ and digested for 1 hour. The cover was removed and the volume of the liquid was reduced to a few mL. 50 mL of 1 M ammonium acetate was added in the cool moist residue, following 16-hours shaking at ambient temperature (overnight). No delay should occur between the addition of the extractant solution and the beginning of the shaking. The extract was separated by centrifugation and decanted into a high pressure polyethylene tube. Stoppering and retaining were done as before for analysis.

The residual fraction was calculated through the total heavy metal concentrations and the concentrations from Step I to Step III.

All the samples were determined for all corresponding heavy metals by ICP-OES (Model 2380, Perkin-Elmer, USA). To control the data quality, soil standards of Chinese National Geological Certified Materials, GSS-4 and GSS-5, were determined together with the samples. The analysis results are in agreement with the standard values.

3 Results and discussion

3.1 Total concentrations of heavy metals in surface soils

We made research on the mean concentrations of heavy metals in surface soils of Guiyang City.

Compared to the average concentrations in urban soils in the world (Table 1), the values of Pb and Cd were much lower than those reported for samples from Hong Kong and Warsaw, but they were similar to those for samples from Bangkok City, Thailand. As for the elements Cr, Cu and Zn, their concentrations were almost higher than those of other cities (Table 1).

In comparison with the background values of Guiyang presented in Table 1, we can see that the average concentrations of heavy metals in soils were higher. The reason why soil samples have the highest Cr value in industrial areas was connected with the waste water discharged from refractory plants, Guizhou Aluminium Branch, electroplating and so on. The soil contamination of Cu in Guiyang might be ascribed to the mining of magnesium in the city (Wang Ji, 2004). The pollution of Pb might be connected with the exhaust gas discharged from smelting factories and chemical plants, especially the exhaust gas from smelting factories, which has higher concentrations of Pb and then was deposited in the surficial soil. In addition, most places in the city are near the traffic crowded roads, although petrol with Pb additives has been banned in Guiyang, Pb pollution may continue to affect the soil environment for years to come. Higher Zn concentrations were recognized in transportation areas because Zn was the additive of vehicle tire and the abrasion of vehicle tire could produce particulates including Zn (Guo Ping et al., 2005). The main pollution sources of Cd in Guiyang were smelting, electroplating and battery factories. It was reported that the pollution of Cd in Guiyang has something to do with the development of industry (Deng Qiujing et al., 2006).

Table 1. Comparison of average metal concentrations in urban soils from different cities in the world and background values in Guiyang (mg/kg)

City	Cr	Cu	Pb	Zn	Cd	Reference
Warsaw	32	31	57	166	0.73	(Daniela Salvagio Manta et al., 2002)
Aberdeen	23.9	27	94.4	58.4	——	(Paterson et al., 1996)
Bangkok	26.4	41.7	47.8	118	0.29	(Wilcke et al., 1998)
Hong Kong	——	16.1	89.9	58.8	0.94	(Chen Tongbin et al., 1997)
Guiyang	92.9	51.6	44.1	139.3	0.28	(Present study)
Background value in Guiyang area	81.6	32.6	24.7	114.0	0.12	(Lu Yingang and Wang Gong., 2001; Wang Ji, 2004)

There are positive correlations among Cr, Cu, Pb, Zn and Cd concentrations (Table 2). And this might indicate that the five metals had the same source in Guiyang. Significant positive correlations between organic carbon and Cr, Cu, Pb, Zn and Cd concentrations suggested that those five metals have a strong affinity with organic matter. Cr, Pb, Zn and Cd concentrations were inversely correlated with clay contents and were directly correlated with sand contents. The concentrations of Cu had no significant correlation with the contents of sand and silt. The results were quite different from those for natural soils in which the Cr, Cu, Pb, Zn and Cd concentrations increased with increasing clay contents (Xu Jialin and Yang Jurong, 1995).

3.2 Heavy metal concentrations in different zones

According to the different uses of soil in Guiyang City, we compartmentalized different zones such as transportation areas, industrial areas, farmland, residential areas and urban parks.

The concentrations of heavy metals in different functional zones of Guiyang are presented in Table 3. We can see that differences in the concentrations of heavy metals are significant. Cr concentrations are biggest in industrial areas, and in the other zones the Cr concentrations follow the order of increasing: urban park>transportation area>farmland>residential area. The concentrations of Cu in urban parks and transportation areas were 56.81 and 54.40 mg/kg, respectively, and in the other zones the concentrations of Cu follow the order of farmland>residential area>industrial area. The maximum concentrations of Pb were reported in urban parks, and they follow the order of transportation area>residential area>farmland>industrial area in different functional zones. The maximum concentrations of Zn were also reported in urban parks, and its minimum concentrations in residential areas. The concentrations of Cd in the soils of different functional zones showed little difference. Its maximum concentrations were recognized in industrial areas, and its minimum concentrations in residential areas.

Table 2. Correlation coefficients for Cr, Cu, Pb, Zn and Cd contents and urban soil properties (*n* = 62)

	Organic C	Sand	Silt	Clay	Cr	Cu	Pb	Zn	Cd
Cr	0.384**	0.526**	-0.335*	-0.584**	1				
Cu	0.461**	0.281	0.036	-0.443**	0.519**	1			
Pb	0.456**	0.585**	-0.147	-0.415**	0.426**	0.387**	1		
Zn	0.658**	0.397**	-0.034	-0.433**	0.537**	0.785**	0.668**	1	
Cd	0.485**	0.415**	-0.340*	-0.536**	0.618**	0.666**	0.565**	0.427**	1

* Significant at the 0.05 probability level; ** significant at the 0.01 probability level.

Table 3. The heavy metal concentrations of urban soils in different functional zones in Guiyang (mg/kg)

Functional zone		Cr	Cu	Pb	Zn	Cd
Transportation area	Range	46.1–115.6	19.9–85.6	20.6–119.9	74.1–184.2	0.13–0.38
	Mean±S.D.	87.6±19.3	54.4±18.5	44.7±25.0	141.9±29.4	0.29±0.08
Industrial area	Range	57.3–241.4	22.8–86.5	22.4–50.0	79.1–139.7	0.2–0.38
	Mean±S.D.	118.5±77.2	47.0±22.9	31.8±8.8	117.8±22.4	0.31±0.07
Farmland	Range	46.6–122.6	16.0–102.2	9.2–107.6	51.0–228.9	0.12–0.45
	Mean±S.D.	85.5±26.8	52.4±29.7	34.3±21.9	131.0±53.8	0.28±0.11
Residential area	Range	65.3–89.3	29.6–68.6	32.1–41.3	94.9–121.1	0.22–0.30
	Mean±S.D.	80.5±13.2	47.3±19.8	35.6±5.0	111.1±14.2	0.26±0.04
Urban park	Range	35.5–184.3	22.3–147.9	24.1–174.7	98.3–460.4	0.06–0.34
	Mean±S.D.	92.4±35.5	56.8±36.2	73.±51.0	194.4±99.5	0.27±0.06

In urban parks, human beings are much more and soil trampling is much more serious than in other places. Therefore, the quality of soil in urban parks has a direct influence on human health, especially to the growth of children. In the parks under our study, we have found that the concentrations of Cr, Cu, Pb and Zn are much higher, which might be connected with the complicated sources and long-term accumulation of heavy metals in the soil. In Guiyang, most parks are located near the traffic-crowded roads or industrial areas, so the vehicle exhaust and industrial activities would bring more heavy metals into the soil of urban parks. For example, The Hebin Park of Guiyang lies near Ruijin South Road in the proper of Guiyang and there are quite a number of vehicles passing by there, so the vehicle exhaust could attribute a lot to the higher heavy metals concentrations in the park. Another example is the Baiyun Park of Guiyang which is located near the Guizhou Aluminium Branch and the particulates including heavy metals could be transferred to the park, so the soil of the park has been polluted to some extent. Tam et al. (1987) also reported that heavy metals in soils of parks near the roads had obvious relationships with transportation quantities.

3.3 Heavy metal fractions

The contributions of the easily soluble, reducible, oxidizable and residual fractions of Cr, Cu, Pb, Zn and Cd from 62 soil samples are shown in Fig. 2.

The highest contents of Cr were associated with the residual fraction (92.1%). The result was in line with other investigations (Lu Ying et al., 2003). Easily soluble, reducible and oxidizable fractions account for 0.1%, 0.3% and 7.5%, respectively. The amounts of Cr in each fraction followed the increasing order of residual>oxidizable>reducible>easily soluble fractions.

The highest percentage of Cu was associated with the residual fraction (74.3%) and the lowest with the easily soluble fraction (0.4%). 1.0% and 24.3% of Cu were associated with the reducible and oxidizable fractions, respectively. The percentage of the Cu fraction followed the increasing order of residual>oxidizable>reducible>easily soluble fractions.

Lead in Guiyang soils was mainly associated with the oxidizable and residual fractions, accounting for 45.8% and 41.9%, respectively. The easily soluble fraction was lowest in Pb, only accounting for 1.1%. The percentage of Pb in the reducible fraction was 11.2%. The proportion of Pb fractions followed the increasing order of oxidizable > residual > reducible>easily soluble fractions.

The percentages of Zn in the easily soluble, reducible and oxidizable fractions were 1.1%, 1.7% and 0.4%, respectively. Most of Zn, with an average of 96.8%, was present in the residual fraction. The percentage of Zn fractions followed the increasing order of residual>reducible>easily soluble>oxidizable fractions.

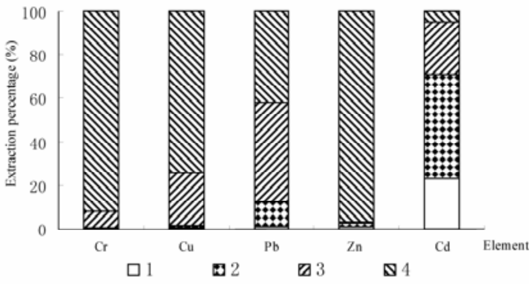

Fig. 2. Contributions of Cr, Cu, Pb, Zn and Cd in the sequentially extracted fractions. 1. Easily soluble fraction; 2. reducible fraction; 3. oxidizable fraction; 4. residual fraction.

The fractions of Cd were quite different from those of the other four elements. The highest contents of Cd were associated with the reducible fraction (47.5%) and the lowest with the residual fraction (5.0%). The amount of Cd in each fraction followed the increasing order of reducible>oxidizable>easily soluble>residual fractions.

The easily soluble fraction was the first to be brought into solution and was considered to be easily available for plant uptake, the reducible fraction was bound mainly with Fe-Mn oxides and it was unstable under low Eh conditions, the oxidizable fraction could be degradated under oxidizing conditions and the residual fraction was not considered to create a bioavailable pool since it was not expected to be solubilized over a reasonable period of time under natural conditions (Lu Ying et al., 2003). The amounts of non-residual fractions represent those of active heavy metals (Xu Jialin and Yang Jurong, 1995). The non-residual fractions of Cr, Cu, Pb, Zn and Cd in Guiyang soils were 7.9%, 25.7%, 58.1%, 3.2% and 95.0%, respectively, which suggested that the mobility and bioavailability of the five metals probably declined in the following order: Cd, Pb, Cu, Cr and Zn. It was known that Cd had extremely high concentrations in the non-residual fraction, which indicated that Cd was more active and it had a potential impact on the environment.

4 Conclusions

In comparison with their background values, the soils in Guiyang have been polluted to different extents by heavy metals. There is a positive correlation between organic carbon and Cr, Cu, Pb, Zn and Cd concentrations. Chromium, Pb, Zn and Cd concentrations are negatively correlated with clay contents and positively correlated with sand contents. The concentrations of Cu show no significant correlation with the contents of sand and silt. The concentrations of Cr, Cu, Pb, Zn and Cd in urban soils of Guiyang vary significantly from one to another. In the parks of Guiyang, the concentrations of Cr, Cu, Pb and Zn are much higher, which might be connected with the complicated sources and long accumulation of heavy metals in the soils.

The distribution of heavy metal species varies greatly among the samples. The metals Cr, Cu and Zn are predominately present in the residual fraction. The concentrations of Cr and Cu in each fraction follow the increasing order of residual > oxidizable > reducible>easily soluble fractions. The amount of Pb in each fraction follows the order of oxidizable>residual>reducible>easily soluble fractions, Zn in each fraction follows the order of residual> reducible>easily soluble>oxidizable fractions and the percentages of Cd fractions follow the order of reducible>oxidizable>easily soluble>residual fractions. The mobility and bioavailability of heavy metals follow the order of Cd>Pb>Cu>Cr>Zn. Some effective measures should be taken to control the mobility and bioavailability of heavy metals in order to protect our environment and the health of human beings.

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